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EFFECT OF BACTERIA AND CARBOXYLIC ACIDS ON REACTIVE SILICA OF BAUXITE ORES

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EFFECT OF BACTERIA AND CARBOXYLIC ACIDS ON REACTIVE SILICA OF BAUXITE ORES

by

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ABSTRACT

Neither organic carboxylic acids (citrate, itaconate, 2-ketogluconate, malate or tartarate) nor microorganisms (Aspergillus niger, Pseudomonas ATCC 21025, Bacillus mucilaginosus strain 0545, species of bacteria isolated from bauxite ores) solubilized reactive silica from bauxite ores and kaolin preparations sufficiently for possible commercial exploitation. Arthrobacter globiformis, A. simplex, A. tumescens, Nocardia farcinica, Azotobacter beijerinckii, and eight unidentified species of bacteria of the genera Arthrobacter and Nocardia were isolated from bauxite ores of Australian, Brazilian and Jamaican deposits.

L'EFFET DES BACTÉRIES ET DES ACIDES CARBOXYLIQUES SUR LA SILICE RÉACTIVE DES MINERAIS DE BAUXITE

par M. Silver

RÉSUMÉ

Ni les acides carboxyliques organiques (le citrate, l'itaconate, le ketogluconate-2, le malate et le tartarate) ni des microorganismes (Aspergillus niger, le Pseudomonas ATCC 21025, le Bacillus mucilaginosus genre 0545, les espèces de bactérie provenant des minéraux de bauxite) n'ont pu solubiliser la silice réactive présente dans les minerais de bauxite et les préparations de kaolin de façon à satisfaire à l'exploitation commerciale possible. On a isolé, des minerais provenant des gisements australiens, brésiliens et jamaiquains, de Arthrobacter globiformis, du A. simplex, du A. tumescens, de la Nocardia farcinica, de Azotobacter beijerinckii, et huit espèces de bactérie non identifées du genre Arthrobacter et Nocardia.

INTRODUCTION

The objective of this project is to examine the feasibility of using microorganisms for the removal of reactive silica from bauxite ores. The most commonly used process for the extraction of aluminum from bauxite ores is the Bayer process, which uses sodium hydroxide. The aluminum containing minerals, gibbsite ($Al_2O_3 \cdot 3H_2O$) or boehmite and diaspore ($Al_2O_3 \cdot H_2O$) are solubilized by this reagent (1) according to the reactions:

$$Al_2O_3 \cdot 3H_2O + 2NaOH \rightarrow 2NaAlO_2 + 4H_2O$$

$$Al_2O_3 \cdot H_2O + 2NaOH \rightarrow 2NaAlO_2 + 2H_2O$$

Impurities in the bauxite ores interfere with this process. The most common and troublesome of these impurities is reactive silica, mostly occurring in the bauxite ores as the mineral kaolinite. Reactive silica is attacked by sodium hydroxide with the formation of an insoluble silicate (1) according to the reaction:

$$Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O + 2NaOH \rightarrow Na_2O \cdot Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O + H_2O$$

Approximately one pound of reagent (sodium hydroxide) and one pound of aluminum are lost for every pound of reactive silica in the bauxite ore (1). The maximum concentration of reactive silica acceptable for the Bayer process is approximately 5% (2). Mechanical processes for the beneficiation of these low-grade (high reactive silica) bauxites are not economic as the recovery of alumina seldom exceeds 70% and the concentration of alumina in the bauxites increases by less than 7%. The reactive silica is finely disseminated in the bauxite ores, and many ores are unsuitable for mechanical beneficiation treatment. For the recovery of aluminum from these ores, complex and expensive technology, such as the sinter process must be used (3). Thus, an economic process for the removal of reactive silica from bauxite ores would be highly beneficial in that much of the world's low-grade (high silica) bauxites would become economically exploitable. For this purpose, a method employing bacteria

was proposed, and the research to 1975 on microbial destruction of silicates was reviewed (3).

Since 1975, Russian and Bulgarian scientists have published reports on the removal of reactive silica from bauxite ores (3,4,5,6,7,8,9). Using microorganisms, called the "silicate bacteria", Bacillus mucilaginosus, 50-70% of the reactive silica could be removed by solubilization while less than 20% of the alumina was solubilized and hence lost from bauxite ores (3). Augmenting the media with glucose or other organic carbon sources (4,6,) and replacing the leaching media at periodic intervals increased silica removal from 45 to 70%. Groudev and Genchev (5) found that cultures of silicate bacteria modified in the laboratory were more effective than wild-type bacteria. The use of simple leaching media (7) and the effects of metal ions (9) showed that this process could be used with ease and without complications for the beneficiation of high - silica bauxite ores.

ALCAN is a Canadian-based international company that produces alumina from bauxite ores. Amongst these are bauxite ores of high reactive silica content, the upgrading of which, by mechanical or chemical procedures, is uneconomical. The dissolution of reactive silicate appears to be an economically attractive alternative. In an attempt to duplicate the results of the Eastern European investigators in which as much as 75% of the silica was extracted from bauxite ores, CANMET undertook to investigate the action of a variety of microorganisms on high-reactive silica-containing bauxite ores.

EXPERIMENTAL

TEST MATERIALS

To evaluate the effectiveness of microorganisms on the dissolution of reactive silica, the following test materials were used: Anachemia kaolin; kaolin clay prepared from ALCAN Cape York (Australia) bauxite; a very high silica Cape York bauxite; a high silica high carbon Australian bauxite, a high silica low carbon Brazilian bauxite; and a low silica Jamaican bauxite. The chemical compositions of these materials are shown in Table 1.

Table 1 - Elementary composition of test materials (%)

Component	Anachemica Kaolin	ALCAN Kaolin Clay	Cape York Bauxite	Australian Bauxite	Brazilian Bauxite	Jamaican Bauxite
Aluminum	20.90	20.28	27.04	20.41	25.81	25.60
Silicon	21.29	21.36	6.34	3.22	3.08	0.59
Tot. Iron	-	-	5.64	2.91	12.53	13.37
Tot. Carbon	-	-	0.13	0.33	0.04	0.33

CHELATING AGENTS

The action of microorganisms on the dissolution of silicate minerals has been reported to be due their production of chelating agents (6). For this purpose, the effects of citrate, malate, itaconate, tartarate and 2-keto gluconic acid on the solubilization of silica from kaolin were examined.

MICROORGANISMS

The following microorganisms were obtained from culture collections: Aspergillus niger, Pseudomonas ATCC 21025 and Bacillus muciloginosus.

Aspergillus niger: The A. niger culture was obtained from the National Research Council of Canada Culture Collection, Ottawa, and carried the identification number NRC - 401128. This culture was formerly identified as strain A-1-215 which produced citric acid as the only organic acid end product; yields were equivalent to 70% sugar conversion (10). It has been deposited with the American Type Culture Collection, to which the catalogue number ATCC 10577 was assigned.

Pseudomonas ATCC 21025: Duff and Webley (11) and Webley, Duff and Mitchell (12) showed that an unidentified Pseudomonas sp. could degrade a variety of synthetic calcium, zinc and magnesium silicates, a variety of natural silicate minerals, namely wollastonite ($CaSiO_3$), chabazite ($CaAl_2SiO_4O_{12}\cdot 6H_2O$), calcium montmorillonite ($Ca(Al,Mg)_6Si_{12}O_{30}(OH)_6\cdot H_2O$), xonolite ($Ca_6Si_6O_{18}\cdot H_2O$), apophyllite ($KCa_4FSi_4O_{10}\cdot 8H_2O$) and olivine ($(MgFe)_2SiO_4$), and number of synthetic minerals, namely tobermorite ($Ca_4Si_5O_{14}\cdot xH_2O$) and hydroxyapatite ($Ca_{10}(OH)_2(PO_4)_6$). This organism, which had been deposited with the American Type Culture Collection and given the catalogue number ATCC 21025, was purchased.

Bacillus mucilaginosus: A culture was obtained from the German Collection of Microorganisms (Deutsche Sammlung von Mikroorganismen), Gottingen, Germany, that had been deposited under the name of Bacillus mucilaginosus by the Institute of Microbiology, USSR Academy of Sciences, Moscow. This culture had originally contained two different species of bacteria, only one of which formed the mucoid colonies characteristic of this species. As this culture did not form spores and were smaller in dimension then described (13), there is considerable doubt that this culture is authentic (14). All other attempts to obtain an authentic culture of B. mucilaginosus from Eastern Europe were unsuccessful. The culture used in this study is known as Bacillus mucelaginosus 0545 by the German Collection of Microorganisms.

MICROORGANISMS ISOLATED FROM BAUXITE ORES: Bacteria were isolated from fresh Australian, Jamaican and Brazilian bauxite ores by the application of a slurry of these ores on the surface of Petri dishes containing the solid medium of Bunt and Rovira (15). Colonies of these bacteria were transferred to Trypticase Soy Broth, and identified according to morphological and biochemical characteristics described in Bergey's Manual of Determinative Bacteriology (16).

MEDIA

Unless otherwise indicated, the solubilization of reactive silica was investigated using the following media: for A. niger - Moyer (17); for B. mucilaginosus - Tesic and Todorovic (13); for Pseudomonas ATCC 21025 - Bunt

and Rovira (15); for the bacteria isolated from the bauxite ores - Trypticase Soy Broth. The compositions of all these media are described in the Appendix.

LEACHING METHODS

Routinely, the leaching of bauxite ores or kaolin clays was accomplished by treating these materials as a 20% slurry with 50 or 100 mL of leachant in Erlenmeyer flasks of 250 mL capacity. The flasks were incubated at 30°C for one week, either on a gyrorotatory shaker or statically with intermittant shaking. To repeat the experimental procedure of Andreev, Polkin, Shavlo, Karavaiko and Morozova (3), 10 g of test material was combined with 7 mL of autoclaved water to form a paste, to which 25 mL of a bacterial suspension was added and incubated at 30°C for 7 days with intermittant shaking. To repeat the procedure of Andreev, Lycheva and Segodina (6), 50 g of test material was suspended in 500 mL of distilled water and bacterial suspension, with or without 1% (w/v) glucose, and incubated at 30°C for 7 days with intermittant shaking. The liquid phase from each of these tests was recovered by filtration and the silicon concentration was determined by atomic absorption spectrophotometry.

RESULTS

EFFECT OF CHELATING AGENTS

The results (Table 2) of leaching tests in the presence of caboxylic acids that can be produced by microbes show that these compounds alone are not able to extract silicon from kaolin clay.

Table 2 - Effect of carboxylic acids on the extraction of silicon from ALCAN kaolin clay

Chelating Agent (Concentration)	Concentration of Silicon in Solution (g/L)
None (control)	0.004
Citrate (42 g/L)	0.010
Citrate (84 g/L)	0.014
Malate (1 g/L)	0.004
Itaconate (1 g/L)	0.004
2-ketogluconate (1 g/L)	0.004
Tartarate (1 g/L)	0.008

The concentration of kaolin was 2 g/100 mL in distilled water. Incubation was at 30°C for 7 days with intermittant shaking. 100% extraction = 4.26 g silicon/L.

As the concentration of silicon in the presence of reagents was near or only slightly above the concentration of the solubility of aluminum silicates in water (0.005 g/L), the conclusion can be drawn that carboxylic acids alone are not capable of extracting silicon from kaolin.

EFFECT OF THE MICROORGANISMS A. NIGER, B. MUCILAGINOSUS AND PSEUDOMONAS ATCC 21025 ON THE SOLUBILIZATION OF SILICON

The leaching of either Anachemia kaolin or ALCAN kaolin clay with the above microorganisms resulted in the extraction of silicon to yield concentrations of between 0.008 and 0.014 g/L. The results were similar whether the flasks were shaken constantly or intermittantly.

The experimental procedure of Andreev, Polkin, Shavlo, Karavaiko and Morozova (3) was repeated with B. muciloginosus and with Pseudomonas ATCC 21025 using the Cape York bauxite. The concentration of silicon released into solution in these leaching tests were 0.010 g/L in the presence of B. mucilaginosus and 0.030 g/L in the presence of Pseudomonas ATCC 21025.

The experimental procedure of Andreev, Lycheva and Segodina (6) was repeated with B. mucilaginosus and with Pseudomonas ATCC 21025 using Cape York bauxite with or without glucose. B. mucilaginosus solubilized silicon to a concentration of 0.016 g/L in the presence of glucose and 0.010 g/L in the absence of glucose. Pseudomonas ATCC 21025 solubilized silicon to a concentration of 0.010 g/L in the presence of glucose and 0.006 g/L in the absence of glucose.

The maximum concentrations of silicon in solution that results from the above experiments are approximately six times the solubility concentration of the silicon of silicate minerals (0.005 g/L). This is insufficient for commercial exploitation. Less than 1% of the silicon was extracted in any of the above leaching tests compared to 34-36.5% (3) and 45-65% (6) reported previously. As the results obtained in this investigation were at variance with those of the Russian investigators, the possibility exists that the optimum microorganisms were not being used.

ISOLATION AND IDENTIFICATION OF MICROORGANISMS FROM BAUXITE ORES

The identification of bacteria in the bauxite ores from Australia, Jamaica and Brazil are shown in Table 3.

Table 3 - Identification of bacteria in Australian, Jamaican and Brazilian bauzite ores

Bauxite Ore	Species Identified
Australian	*Nocardia farcinica* *Arthrobacter globiformis* *Arthrobacter tumescens*
Jamaican	*Arthrobacter tumescens* *Arthrobacter globiformis* *Arthrobacter* spp. strains designated 4J, 6JS, 6JL, and 16J *Nocardia* spp., strains designated 2J and 5J2
Brazilian	*Arthrobactor globiformis* *Arthrobacter simplex* *Azotobacter beijerinckii* *Nocardia* spp., strains designated 10T and 11T

These bacteria were identified according to the characteristics listed in Bergy's Manual of Determinative Bacteriology, 7th edition (6). Where insufficient characteristics of the isolated bacteria match the description of known and described species, the bacteria are identified only to the genus level.

EFFECT OF MICROORGANISMS ISOLATED FROM BAUXITE ORES ON THE SOLUBILIZATION OF SILICON

None of the bacteria isolated from the bauxite ores could be grown in a slurry of any of the bauxite ores in water supplimented with glucose. As Trypticase Soy Broth was suitable for abundant growth of all isolated bacterial strains, this medium was used to test the abilities of these bacteria to solubilize silicon from all of the bauxite ores and kaolin clay supplied by ALCAN. None of these bacteria were able to release silicon into solution to concentration greater than 0.044 g/L from any of the bauxite ores or the kaolin clays described in Table 2.

DISCUSSION

The results of this study show that none of the carboxylic acids nor any of the microorganisms employed solubilized silicon from any of the bauxite ores or kaolin preparations to the extent required for commercial exploitation. This may be due to any one or a combination of the following factors:

1. The reactive silica in the bauxite ores or the kaolin preparations may be resistant to the extractive procedures used.
2. The solubilization agents (chelating agents or microorganisms) used may be ineffective in the solubilization of silica.
3. The procedures used may not have been optimum.

Differences in the extraction of components, including silica, from various clay raw materials (kaolin) were reported by Vainberg et al. (18), who were able to solubilize 6 to 8% of the silica without replacement of the leaching media and 16 to 26% of the silica with replacement of media. Groudev and Genchev (5) were able to solubilize 26 to 64% of the silica from two different bauxites; Andreev, Polkin, Shavlo, Karavaiko and Morozova (3) were able to solubilize 23 and 50% of the silica from two different bauxites; and Andreev, Lycheva and Segodina (6) were able to solubilize 40 to 58% of the silica from four different bauxites. Thus, differences are noted between the extents of the extractions of silica from mineral preparations that are amenable to bacteriological leaching; no mention is made of ores that are resistant to these procedures.

The involvement of organic chelating agents of microbial origin is suggested by Andreev, Polkin, Shavlo, Karavaiko and Morozova (3) and Groudev and Genchev (5) to influence silicon solubility from 0.5 to 5 fold, while strongly complexing agents increase silicon dissolution up to 50 fold. Huang and Keller (19) were able to solubilize silicon to concentrations of approximately 0.01 g/L from a variety of American kaolin clays by carboxylic acids. Groudev and Genchev (5) found that cultures of the "silicate bacteria" modified in the laboratory were more effective in the solubilization of silicon than wild-type cultures, and that

microorganisms other than the "silicate bacteria", while increasing the solubility of silica in aqueous solution, were less effective. Thus, not only the species but also the strain of bacteria employed appear to be of critical importance in the solubilization of silicon from kaolin and other reactive silica-containing materials. The most probable reason that silicon was not solubilized in the tests of this investigation is that microorganisms suitable for this procedure were not available.

The microorganims isolated from the Australian, Brazilian and Jamaican bauxites are predominantly of the genera Arthrobacter and Nocardia. These are obligate aerobic filamentous bacteria that are common in soils (16). The Brazilian bauxite also contained Azotobacter beijerinckii, a nitrogen-fixing bacterium. Karavaiko et al. (20) identified a number of species belonging to the genera Arthrobacter, Nocardia and Pseudomonas and nitrogen-fixing bacteria in Russian spodumeme ($LiAlSi_2O_6$) deposits. These bacteria, various fungi and "silicate bacteria" (21) participate in the degradation of spodumeme. In alkaline and neutral conditions, montmorillonite is formed according to the reaction

$$2\ LiAlSi_2O_6 + 2\ H_2O \rightarrow Al_2(Si_4O_{10})(OH)_2 + LiOH$$

In acidic conditions, montmorillonite is converted to kaolin according to the reaction

$$2\ Al_2(Si_4O_{10})(OH)_2 + 2\ LiOH + 5\ H_2O + CO_2 \rightarrow Al_4(Si_4O_{10})(OH)_8 + Li_2CO_3 + 4\ H_2SiO_3$$

In the presence of sulphate, spodumene is converted to kaolinite according to the reaction

$$4\ LiAlSi_2O_6 + 6\ H_2O + 2\ H_2SO_4 \rightarrow 2\ Li_2SO_4 + Al_4(Si_4O_{10})(OH)_8 + 4\ H_2SiO_3$$

Kaolinite is generally regarded as the final identifiable mineral in the weathering sequence of the silicate minerals, and may also form either by crystalization from aluminum and silicon-rich solutions or by resilication of aluminum-rich minerals (22). Desilication of kaolinite may yield boehmite, diaspore or gibbsite (23). The mechanisms of the formation of these aluminum ore minerals is uncertain, but may involve crystalization from solution or weathering directly or indirectly from silicate minerals

with the intermediate formation of aluminosilicates. These mechanisms may not be mutually exclusive (24), but they are very slow and some are reversible. All are poorly understood, as the correlation of laboratory experiments with field observations are inadequate. Thus, the fundamental natures of chemical reactions concerning aluminum ore minerals and clay minerals remain to be resolved.

CONCLUSIONS

1. Neither organic carboxylic acids (citrate, itaconate, 2-ketogluconate, malate or tartarate) nor microorganisms (Aspergillus niger, Pseudomonas ATCC 21025, Bacillus mucilaginosus strain 0545, species of bacteria isolated from bauxite ores) solubilized reactive silica sufficiently for possible commercial exploitation. The possibility exists that the optimum microorganisms for this process were not used.

2. Australian, Brazilian and Jamaican bauxite ores contained bacteria identified as Arthrobacter globiformis, A. simplex, A. tumescens, Nocardia farcinica, Azotobacter beijerinckii, and unidentified species of bacteria belonging to the genera Arthrobacter and Nocardia. However, none of these were able to solubilize silica.

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APPENDIX: MEDIA USED FOR THE CULTIVATION OF MICROORGANISMS

Aspergillus niger: The medium used for the cultivation of A. niger is a modification of the medium described in the footnote of Table 5 of Moyer (17) and contains (in g/L) glucose (120), NH_4NO_3 (2), KCl (0.1), KH_2PO_4 (0.25), $MgSO_4 \cdot 6H_2O$ (0.2), $FeSO_4$ (0.001), yeast extract (0.2) and methanol (30). The medium (minus the glucose and methanol), which was adjusted to pH 5.5 with HCl and glucose in solution were sterilized separately by autoclaving at 121°C for 30 min. The sterilized medium, sterile glucose solution and methanol were combined immediately before use.

Bacillus mucilaginosus: The medium used for the cultivation of B. mucilaginosus is described by Tesic and Todorovic (13) and contains (in g/L) glucose (5), $(NH_4)_2SO_4$ (1), K_2HPO_4 (1), $MgSO_4 \cdot 6H_2O$ (0.5), $FeSO_4$ (0.001). The mineral salts and the glucose were sterilized separately by autoclaving at 121°C for 30 min and were combined immediately before use. The initial pH of the medium was 7.8.

Pseudomonas ATCC 21025: The medium used for the cultivation of Pseudomonas ATCC 21025 is described by Bunt and Rovira (15) and contains (in g/L) K_2HPO_3 (0.4), $(NH_4)_2HPO_4$ (0.5), $MgSO_4 \cdot 6H_2O$ (0.05), $MgCl_2$ (0.1), $FeCl_3$ (0.01), $CaCl_2$ (0.1), Peptone (1), yeast extract (1), soil extract (250). The pH of the medium was adjusted to 7.4 with HCl and sterilized by autoclaving for 30 min at 121°C. For solid medium, agar (15) was added and dissolved by heating.

Bacteria isolated from bauxite ores: The medium used for the cultivation of the bacteria isolated from the bauxite ores was a commercial medium, Trypticase Soy Broth (Becton, Dickinson and Company) which contains (in g/L when prepared) glucose (2.5), K_2HPO_4 (2.5), NaCl (5), phytone peptone (3) and trypticase peptone (17). The pH of the medium is 7.3. Sterilization was by autoclaving at 121°C for 15 min.